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Contractor: Syracuse University

Contract No: DA-CML-18-108-61-G-27

Progress Report No. 5

Covering the Period

June 1, 1961 to August 31, 1961

THE SAMPLING OF AEROSOLS IN A TURBULENT AIR FLOW

Prepared by: V. Goldschmidt

August 31, 1961

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THE SAMPLING OF AEROSOLS IN A TURBULENT AIR FLOW

Prepared by: V. Goldschmidt

August 31, 1961

Approved by: S. Eskinazi

Dr. Salamon Eskinazi
Research Contract
Director

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I. SUMMARY

The use of La Mer generator as an aerosol source for this study has been abandoned. The results of the La Mer generator tested turned out not to be successful. The reasons for attempting to adopt such an aerosol generator was because of it having been initially recommended and provided by the sponsors. However, although the results were not useful to our purpose, the time spent has been very profitable in the overall plan for producing monodispersed aerosols in adequate size and sufficient concentration. During the period covered in this report the variations of output with nucleizer current were observed and noted as too low for the desired test conditions.

Prior to observations of the particle size distribution and mass output from a Vaponefrin nebulizer, an evaluation was made on the spread of liquid drios on a clean glass slide. For sufficiently large drops it appears reasonable to expect a decrease in spread factor with size. However, for droplets between 2 and 30 microns, the spread factor was noted to increase with size. Spread factor is defined as the ratio of the spherical drop diameter to that of the same drop when spread. This number, as defined, is always less than unity. The variation of spread factor with drop size, for the range expected in our work, has been well defined for gravitational settling on clean glass slides. The collection of drops may be made at different speeds, as in our wind tunnel. An evaluation of spread factor with impaction speed other than gravitational will be performed in the future.

A small scale wind tunnel is being constructed in order to evaluate the limitations and determine the calibrations of a hot wire anemometer as a counting device in an aerosol stream. By sampling isokinetically through a filter, the count obtained on the hot wire anemometer will be correlated to the true concentration. For the initial runs a nebulizer has been selected as a generator for a dibutyl phthalate aerosol. In spite of the usual large spread on drop size the nebulizer is adaptable due to its small size and relatively high mass output. The output characteristics of a Vaponefrin H. E. Curry all glass nebulizer are shown in Figures 8, 9 and 10. The small wind tunnel tested has a 2" diameter test section and flows around 12'/sec. Its small size will allow substantial counts on the hot wire anemometer and negligible errors due to settling of the aerosol on the tunnel walls. These are all preliminary measurements, eventually the same tests will be conducted in the full size tunnel described in Progress Report No. 2.

II. THE WORK ACCOMPLISHED DURING THIS PERIOD

1) La Mer Generator

In Progress Report No. 4 the results of extensive studies on the La Mer generator with triphenyl phosphate condensing on sodium chloride nuclei were shown. Shortly after the last progress report an evaluation was made of the variation of output with nucleizer current. It was noted that when poorly filtered and inadequately dried air was used, the output increased with decrease in currents below 1.5 amps. This clearly indicated the presence of impurities in the air which acted as condensation nuclei. As the current increased from zero, the air temperature carrying these impurities also increased. The foreign condensation nuclei would consequently increase in temperature and not allow as much condensation of triphenyl phosphate. The net effect would be a decrease in output until the production of sodium chloride nuclei would be such as to overshadow the effects of the impurities and moisture in the air. In Figure 1 the variations in output with nucleizer current using both air and bottled nitrogen are shown. The drop in output, around 6 amperes, coincides with the nichrome wire coil becoming red hot. (This may correspond to the melting point of NaCl.) The air was filtered through a filter capsule consisting of pyrex wool filtering fibre stuffed with anhydrous $CuSO_4$ into a $1\frac{3}{4}$" x 6" section of pipe and through a second filtering unit consisting of two Gelman type

AM-4 absolute filter pads. The nitrogen was filtered through the AM-4 filter pads (average pore size .85 microns) and through a filter capsule without any $CuSO_4$. (The manufacturer guarantees a moisture content under 5 grains per thousand cubic feet of nitrogen gas.) The maximum output obtainable was above 300 micrograms/liter, or around .002 grams/minute, well below the minimum desirable output of .02 grams/minute as per page 17 of Progress Report No. 4.

The change in mass output by increasing the bubbler flow from 1.6 liter/minute to 3 liter/minute with a nuclizer current of 4.5 amps was an increase by a factor of two. With bubbler flows increasing from 3 to 4.5 liters/minute no substantial increase of output was noted.

CONCLUSIONS

The La Mer generator as tested is not applicable to our use. Its evaluation and consideration was undertaken at the conception of our contract and its use as an aerosol generator was proposed by our sponsors. The particular device on which our tests were made was obtained through the Army Chemical Corps. The La Mer generator was found to be unreliable for duplicating conditions of reproduction of aerosols for the following reasons:

a) The aging of the filament and consumption of NaCl in the nucleizer. Lodge and Tufts (Journal Colloid Science, 1958) have reported these effects.

b) The influence upon the output by the cleanliness of the glass ware has caused some investigators to tear down the

instrument even daily for cleaning. The decomposition of the triphenyl phosphate at high boiler temperatures also necessitates regular fresh starts.

c) The present location of the thermometers in the boiler and the heater are not representative. Their reading has some indication of the temperature levels in the two beakers, but not necessarily to a simple proportion. The thermometer in the heater unit measures the convective currents from the heater unit rather than from the beaker itself.

d) The effect of condensation of triphenyl phosphate on moisture and impurities is noticed at low current levels unless careful drying the filtering is done.

Even if time could be spent to make this a reliable instrument by changing some of the controlling and sensing devices, its relatively low output and small aerosol sizes make its use undesirable. The use of the La Mer generator is dropped in preference to the dust cloud and atomization devices.

of the matrix. Immediately after a sample is collected the coverslip is carefully placed over the slide and, if heated to the proper level, the droplets will be drawn up to their original spherical form as the matrix around them melts and then solidifies. The coverslip must be handled with care and at the right temperature to keep the droplet from deforming and yet allowing it to assume its original spherical shape. Some experimenters, such as Maybank and Fenrick, Reference 3, have been able to freeze the spherical drops and measure their diameter while still frozen.

b) Magnesium oxide method. The pits resulting when droplets hit on a slide coated with the deposit of a burning magnesium ribbon can be measured and correlated to the original size of the droplet. K. R. May, found that for drops between 20 and 200 microns the ratio of true drop size to impression size has an average value of .86 (the values ranged however between .78 and .89). For drops under 10 microns the method is unreliable and hence not directly applicable for the size droplets we expect to encounter.

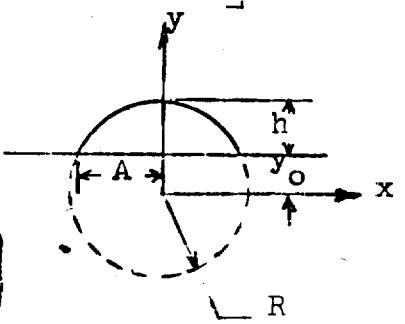
c) Stain method. Droplets may be detected by coating the sampling slide with a medium which either by chemical or physical change gives a stain or mark where the droplets have hit. This method is used mostly for counting rather than sizing.

d) **Plain glass method.** For aerosols with low volatility plain glass slides may be used and microscopic observations made before significant evaporation takes place. By cleaning glass slides thoroughly and polishing them with lens paper after rinsing in aerosol O.T. the droplets will spread un-uniformly and take up equilibrium in some non-spherical form. The ratio of the original drop diameter to the diameter of the spread drop has been defined as spread factor. We shall denote spread factor as s.f., the original drop diameter as d = 2r, and the spread drop diamater as 2A.

If we assume, with May (Reference 2) that the drop will take the form of a spherical segment with radius R, then we can correlate A, R and r.

The volume of the drop based on the spherical drop diameter will be given as $\frac{4}{3}\pi r^3$, whereas the volume of the spherical segment as shown will be given

$$\int_{y_o}^{R} \pi x^2 dy = \int_{R^2-A^2}^{R} \pi(R^2-y^2)dy = \frac{\pi R^3}{3}\left[2 - (2 + \frac{A^2}{R^2})\sqrt{1 - \frac{A^2}{R^2}}\right]$$



Then, as

$$\frac{4}{3}\pi r^3 = \frac{\pi R^3}{3}\left[2 - (2 + (\frac{A}{R})^2\sqrt{1 - \frac{A^2}{R^2}}\right]$$

if we let $\frac{A}{R} = \sigma$,

$$\frac{r}{R} = \frac{1}{2^{1/3}}\left[1 - (1 + \frac{\sigma^2}{2})\sqrt{1 - \sigma^2}\right]^{1/3} \qquad (1)$$

The focal point of the lens formed by the liquid drop can be located by allowing the liquid lens to reflect some distant object and focusing on its image. Let h be the height of the drop, and f the distance to the focal point. The distance from the glass slide to the focal point, will be given as z = h + f, and is a measurable parameter. The focal point is related to R by the index of refraction, η as

$$\frac{1}{f} = (\eta - 1)\frac{1}{R} \qquad (2)$$

and η is a constant depending on the liquid observed.

The height h is related by the geometry of the drop, to the radius R as follows:

$$h = R - \sqrt{R^2 - A^2} \quad ,$$

or

$$\frac{h}{R} = 1 - \sqrt{1 - \left(\frac{A}{R}\right)^2} \qquad (3)$$

As z = h + f we can combine Eqs. (2) and (3) to obtain:

$$\frac{Z}{R} = \mu - \sqrt{1 - \frac{A^2}{R^2}} \qquad (4)$$

where

$$\mu = \frac{\eta}{\eta - 1} \qquad (5)$$

From Eq. (4), and as $\mu > 1$,

$$R = \frac{Z\mu + \sqrt{Z^2 + A^2(\mu^2-1)}}{\mu^2 - 1} \tag{6}$$

As $s \cdot f = \frac{r}{A} = \frac{r}{R} \cdot \frac{R}{A} = \frac{r}{R} \cdot \frac{1}{\sigma}$, from Eq. (1),

$$s \cdot f = \frac{r}{A} = \frac{1}{\sigma}\left\{\frac{1}{2}\left[1 - (1 + \frac{\sigma^2}{2})\sqrt{1-\sigma^2}\right]\right\}^{1/3} \tag{7}$$

where $\sigma = \frac{A}{R}$

The value of s·f can be obtained with only z and A being measured.

For dibutyl phthalate, a plasticizer used as an aerosol source due to its low volatility, the index of refraction is of 1.4925 (Eastman Plasticizers Catalog No. L-104). If $\eta = 1.4925$, then $\mu = 3.03$ and $\mu^2 = 9.18$. Equation (6) becomes

$$R = .37Z + .122\sqrt{Z^2 - 8.183A^2}$$

or

$$\frac{R}{A} = \frac{1}{\sigma} = .37\left(\frac{Z}{A}\right) + .122\sqrt{\frac{Z^2}{A^2} - 8.183} \tag{8}$$

This relationship is shown in Figure 2. In Figure 3 Eq. (7) is plotted. By combining the above curves, one with σ as a function of $\frac{Z}{A}$, the other with spread factor as a function of σ, we obtain the spread factor as a function of Z and A only, as per the curve on Figure 4.

This last relationship enables us, assuming that the drop takes a plano-convex form, to evaluate the spread factor once the spread drop radius and distance from glass slide to the focal point of the lens formed by the droplet are measured.

The spray from a Vaponefrin all glass nebulizer (H. E. Curry, Seattle, Washington) was collected by gravitational settling on a glass slide thoroughly cleaned and polished with aerosol O. T. and lens paper. The resulting dibutyl phthalate drops were observed under the microscope and by measuring the drop diameters and the distance from the glass slide to the focal length of the droplets, the following resulted:

TABLE I

2A-Spread Diameter	Length-Z	Z/2A	s.f. From Fig. 4
3.5	13.0	3.69	.378
4.2	13.7	3.24	.395
4.2	9	2.13	.475
4.9	17.5	3.55	.385
4.9	15.0	3.04	.405
5.6	17	3.02	.407
6.3	18	2.84	.418
6.3	18	2.84	.418
7.0	18.8	2.67	.428
7.0	18.8	2.67	.428

Table I (continued)

2A-Spread Diameter	Length-Z	Z/2A	s.f. From Fig. 4
7.0	19	2.70	.425
7.7	19.8	2.56	.435
9.2	29	3.16	.400
10.6	23	2.18	.470
11.3	30.5	2.71	.425
11.3	28	2.49	.440
12.0	27	2.26	.460
12.0	26	2.17	.470
12.7	35	2.76	.420
13.4	30.3	2.26	.460
14.8	38.2	2.58	.435
17.6	39	2.22	.465
18.3	43	2.35	.451
21.1	45.8	2.17	.470
22.5	41	1.81	.525
26.7	45	1.68	.555
29.6	51.3	1.73	.544
31.68	53.2	1.68	.555
45.1	78.5	1.74	.540
45.7	80.8	1.76	.535
66.18	112.	1.69	.550

The above values are plotted in Figure 5. It is noted that with an increase in drop size the spread factor increases. By the definition of spread factor, as its value increases the amount of spread decreases. For drops in between 1.5 and 35 microns, assuming that their shape when settled on a glass slide is that of a spherical segment, the spread factor increases with drop size and consequently their spread with respect to their original size decreases with increasing size. The data in Figure 5 follows to some approximation the relationship

$$s \cdot f = \frac{r}{A} = .306\ (2A)^{.162} \tag{9}$$

or

$$\frac{A}{r} = \frac{3.27}{(2A)^{.162}} = 2.75\ \left(\frac{r}{A}\right)^{.162}\ r^{-.162}$$

hence

$$\frac{A}{r} = (2.75\ r^{-.162})\ \frac{1}{1.162}$$

and

$$\frac{A}{r} \doteq 2.38\ r^{-.14} \tag{10}$$

The assumption of a spherical shape in the free surface is justifiable as the gravitational forces are negligible compared to the surface pressure, $2\sigma/R$.

In Progress Report No. 4 it was commented that the spread of droplets was expected to be proportional to the original diameter. For

sufficiently large drops, the height reached will be approximately constant from drop to drop. The volume of the spread drop will be given by $\pi A^2 h$, hence

$$\frac{4}{3}\pi r^3 = \pi A^2 h$$

and

$$s.f = \frac{r}{A} = \left(\frac{3h}{4}\right)^{1/3} A^{-1/3}$$

The results obtained indicate however that $s.f \propto A^{.16}$ and hence the assumption of small change of height from drop to drop is not justified. The height, or capillary rise, a, could be approximated for sufficiently large drops as

$$a = \sqrt{\frac{\sigma}{\rho g}}$$

where σ is surface tension.

The actual value of surface tension for dibutyl phthalate is not listed in the typical reference tables, but it is expected to be around 20 dyne/cm. Hence, as $\rho = 1.048$ gm/cc, then $a > .1$ cm which is much larger than the range being observed, and disproves the assumption of slowly varying spread drop height.

The data of Table I is plotted on linear paper in Figure 6, confirming that for drops between 1.5 and 35 microns the spread factor increases with size. For larger droplets there is not sufficient data to determine the relationship of spread factor to size, but it is possible that after a certain size the spread factor might decrease with an increase in diameter.

3) Hot Wire Anemometer For Measuring Aerosol Concentration

As discussed in Progress Report No. 3 and shown in Progress Report No. 4 it is intended to evaluate the adaptation of a hot wire anemometer as a sampling device for an aerosol.

A Vaponefrin nebulizer, "H. E. Curry all glass nebulizer," is being used as an aerosol generator and a small scale wind tunnel is being designed for preliminary runs. The use of a small scale tunnel, with velocities around 10'/sec. at a test section of nominally 2 inch diameter, is being adopted. This size wind tunnel will allow the number of particles impinging on the hot wire anemometer to be of a significant magnitude so that the corrections for deposition and settling of droplets are negligible. The construction of the wind tunnel has been completed and evaluations of its velocity profile are presently being conducted.

The Vaponefrin nebulizer has been selected for preliminary runs as its small dimensions and yet substantial output make it easily adaptable to the small wind tunnel. A schematic of the nebulizer is shown in Figure 7. A stream of air blowing through the nozzle causes a suction on the dibutyl-phthalate in the capillary tube. When the air flow is of sufficient magnitude the dibutyl-phthalate flows over the capillary tube and breaks up into smaller drops as it is hit by the air blast. The baffle stops the larger droplets and a relatively homogeneous spray of dibutly-phthalate results. The output is depend-

ent on the air flow and the distance from the nozzle to the liquid level (and hence amount of liquid in the nebulizer). Through the port located above the nozzle liquid may be fed at the same rate it is being sprayed and a steady state condition can be reached. Figure 8 shows the relationship of output to air flow and volume of dibutyl phthalate in the nebulizer. At air flows around 4.5 liters/minute an upstream pressure if 15 psi was noted, and sonic velocities were reached at the nozzle. This stage coincides with a change of slope in the output curves.

The particle size distribution was observed at an air flow of 4.45 liters/minute and with 6 ml of dibutyl phthalate in the nebulizer. The sample was collected on a clean, polished glass slide as follows:

> The nebulizer was centrally placed in an exhaust hood approximately 22" high by 30" wide and 19" deep. The glass slide was placed on the base of the hood approximately 12" ahead of the nebulizer. After running for one minute the exhaust fan on the ceiling of the hood was disconnected. The capacity of the fan is such that during its operation no particles were noticed to settle on the glass slide. The nebulizer operated for 60 more seconds after which the hood was maintained closed for more than 10 minutes allowing the spray to settle gravitationally.

The spray collected on the glass slide was observed by two operators and the following noted:

	Operator 1	Operator 2
Arithmetic Mean	5.97	5.60
Standard Deviation	4.27	4.30
Mean Surface Dia.	7.34	7.22
Mean Volume Dia.	8.76	9.08
Total Number of Counts	449	412

Figure 9 shows the probability curve and Figure 10 shows the frequency distribution curve corresponding to the second column of data.

In order to evaluate whether the location of the slide with respect to the nebulizer had a direct influence in favoring particles by their size, a similar test was repeated placing one slide approximately 12" ahead of the nebulizer, a second right under the nebulizer and a third approximately 5" behind it. All slides were on the base of the hood, and the nebulizer was still centrally placed approximately 10" above the slides. The following was noted:

	Slide Ahead	Slide Under	Slide Behind	All Three
Arithmetic Mean	4.23	7.63	5.84	5.65
Standard Deviation	3.35	4.3	4.53	4.25
Mean Volume Dia.	6.65	10.02	9.17	8.85
Total Number of Counts	143	93	95	331

The combination of all three slides yields averages in very good agreement with the results obtained by the two different operators in the previous test.

It would have been desirable to obtain a good correlation in between the three slides, but with counts as low as 143, 93 and 95 a correlation would have been coincidental. Construction of an instrument for enclosing an aerosol, such as the output of the above nebulizer, has just been completed. Basically it consists of a cylindrical section through which the aerosol flows undisturbed; upon triggering a spring loaded mechanism a volume within this cylinder is enclosed, and by placing the instrument in a vertical position the particles trapped in it will settle on a glass slide within the enclosed volume. During the following period it will be used to confirm particle size distribution obtained from the Vaponefrin nebulizer.

If the output of the nebulizer is known, in quantity and size distribution, a hot wire anemometer may be placed in the stream in such a manner so the impinging droplets will cause a change in the temperature, and resistance, of the heated wire.

The output of the hot wire anemometer can then be amplified and introduced into an electronic counter sensitive to voltage rises above a certain mean. According to the amplification on the amplifier and the sensitivity on the counter, the voltage changes counted are caused by particles impinging on the wire, turbulence of the air stream or noise in the amplifier. The ideal conditions would be given by an aero-

sol whose blips due to impaction are in excess of the voltage variations due to turbulence. In such a case, by proper discrimination the counter can be set to record only the impactions of the droplets on the wire.

Preliminary runs with a .00018" diameter tungsten wire have been made. Due to the diameter of the fine wire being of the same order of magnitude as the aerosol particles, the impaction efficiency is close to unity. The value of impaction coefficient was calculated with aid of the relationships shown in Reference 4 and for all practical purposes it may be considered as 100 percent.

The expected count on the hot wire anemometer was estimated and the actual count compared to it. In preliminary runs counts in the order of 200 counts/second were obtained, with the measured counts being higher than the estimated values. However, not enough data has been gathered yet to make any valid conclusions. Tests are being run at this moment to further evaluate the use of the hot wire anemometer as an instrument for measuring aerosol concentrations.

III. WORK PLAN FOR THE FOLLOWING PERIOD

In the following period the correlation of counts on the hot wire anemometer to actual concentrations is to be made. The small scale wind tunnel will be improved so a desirable velocity profile is obtained. Measurements of concentration will be calibrated by sampling isokinetically through a nozzle and colorimetrically measuring the sample collected on a filter pad.

The quality of the aerosol and correlations between hot wire results and isokinetic sampling will be improved. An attempt will be made at perturbing the equations of motion for an aerosol in air, giving the important Reynolds type equations for the case of an areosol in a turbulent stream.

REFERENCES

1. K. R. May, - Journal Scientific Instruments, Vol. 27, p. 128, May, 1950.

2. K. R. May, - Journal Scientific Instruments, Vol. 25, p. 187, October, 1949.

3. J. Maybank and W. J. Fenrick, - Suffield Experimental Station, Technical Paper No. 186.

4. W. E. Ranz, - University of Illinois, Engineering Experiment Station, SO-1004, March 31, 1951.

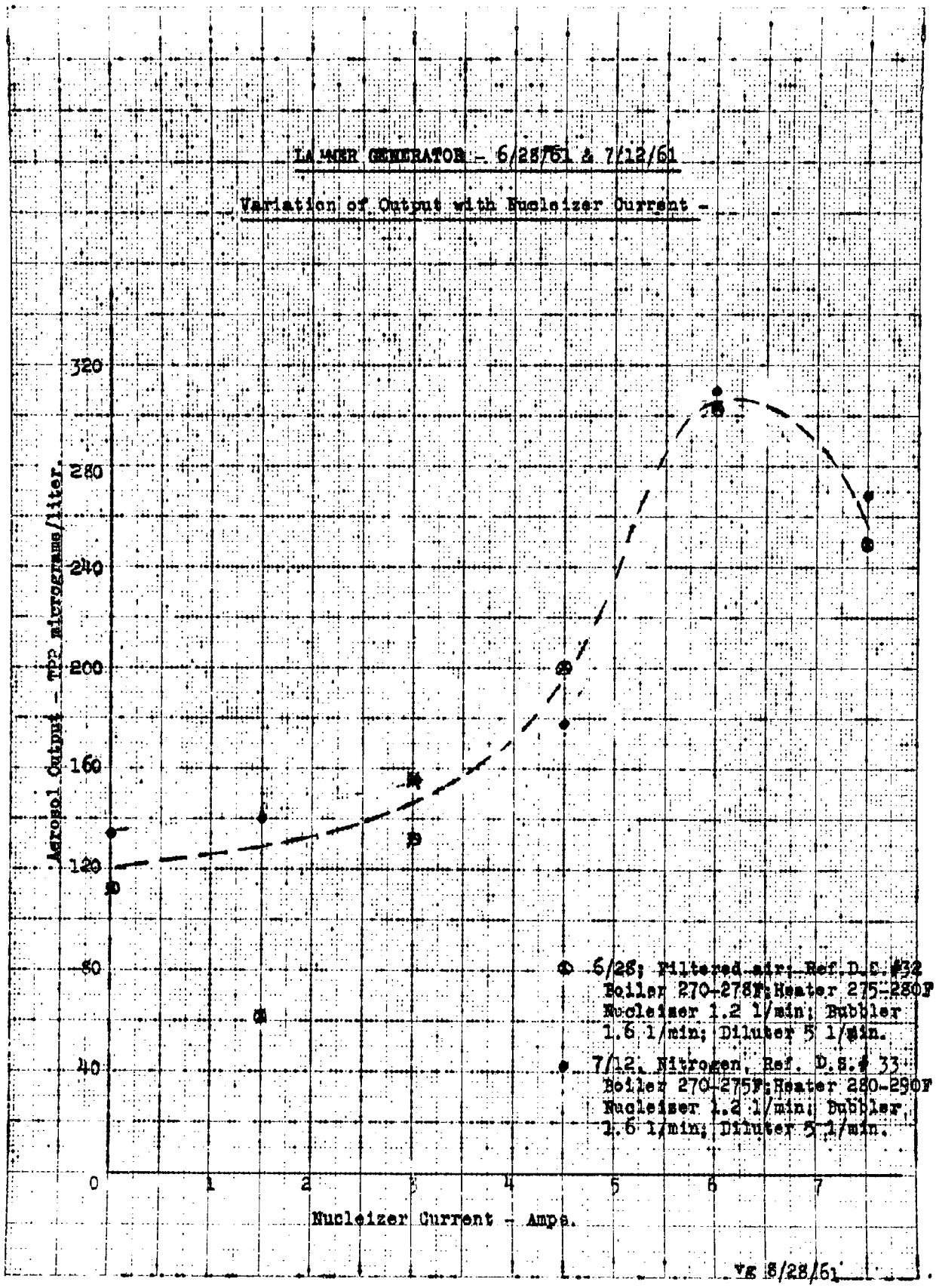

LAMER GENERATOR - 6/28/61 & 7/12/61
Variation of Output with Nucleizer Current -
Aerosol Output - TPP micrograms/liter.
320
280
240
200
160
120
80
40
0
1
2
3
4
5
6
7
Nucleizer Current - Amps.
6/28; Filtered air; Ref. D.S. #32
Boiler 270-278F; Heater 275-280F
Nucleizer 1.2 l/min; Bubbler
1.6 l/min; Diluter 5 l/min.
7/12, Nitrogen, Ref. D.S.# 33
Boiler 270-275F; Heater 280-290F
Nucleizer 1.2 l/min; Bubbler
1.6 l/min; Diluter 5 l/min.
VE 8/28/61

Fig. 1

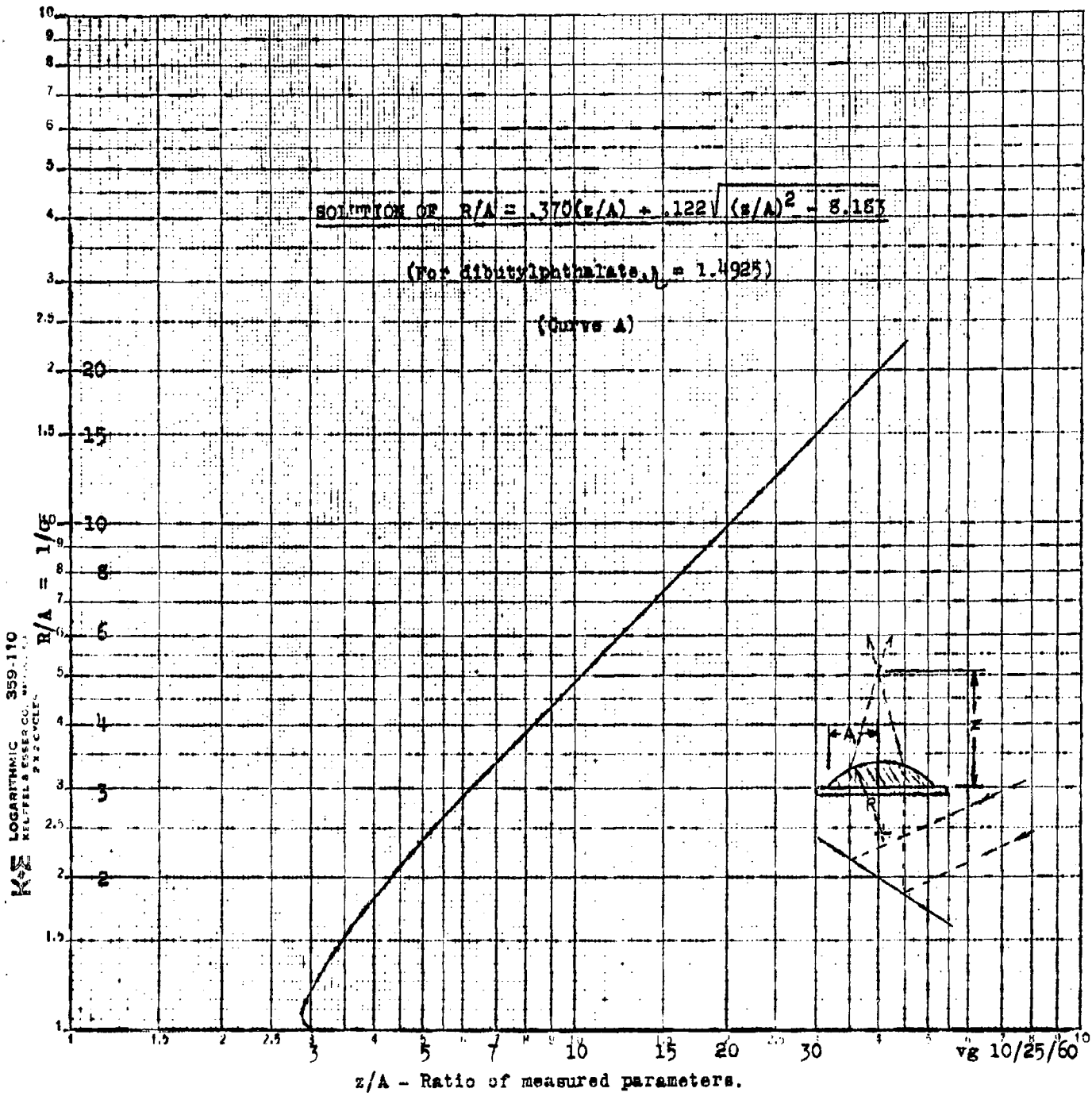

SOLUTION OF R/A = .370(z/A) + .122√(z/A)² - 8.163
(For dibutylphthalate, μ = 1.4925)
(Curve A)
R/A = 1/G
LOGARITHMIC
359-110
z/A - Ratio of measured parameters.
vg 10/25/60

Fig. 2

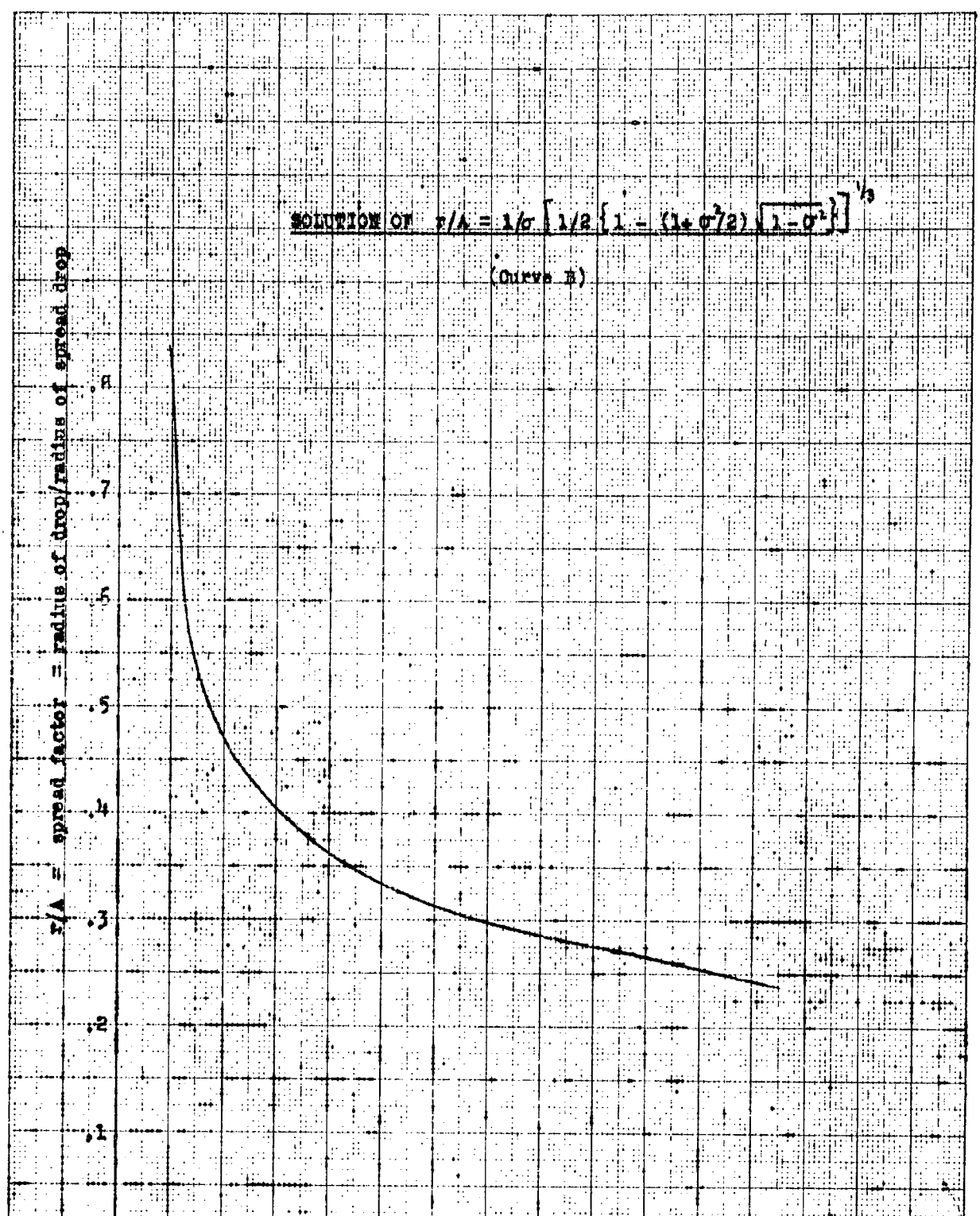


Fig. 3

K&E 10 X 10 TO THE CM. 359-14
KEUFFEL & ESSER CO.

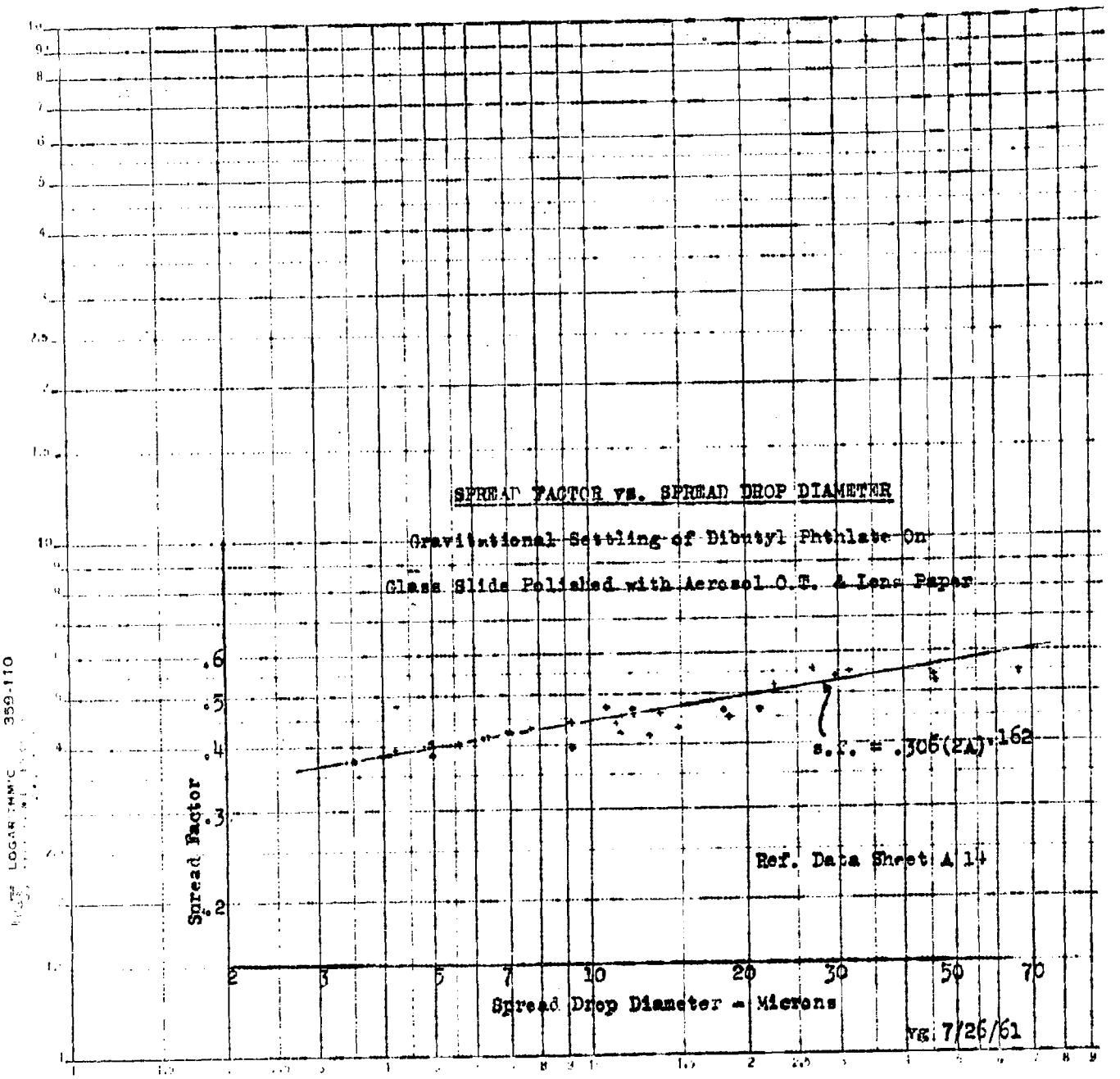
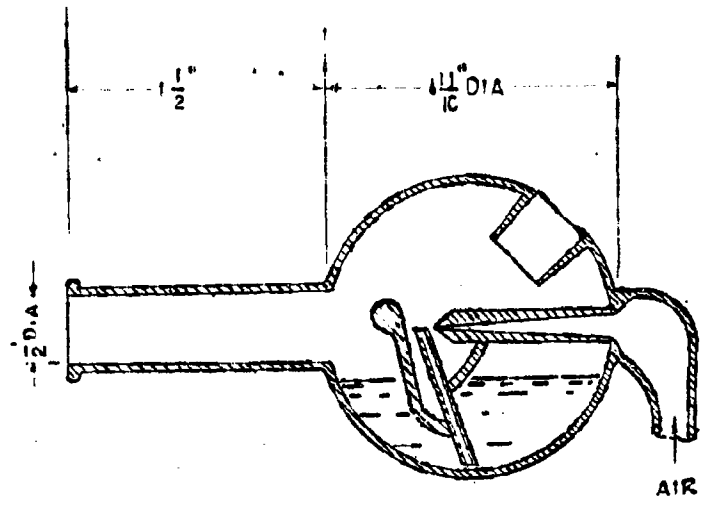

SPREAD FACTOR vs. SPREAD DROP DIAMETER
Gravitational Settling of Dibutyl Phthlate On
Glass Slide Polished with Aerosol O.T. & Lens Paper
S.F. = .306(2A)^.162
Ref. Data Sheet A 14
Spread Factor
.6
.5
.4
.3
.2
2
3
5
7
10
20
30
50
70
Spread Drop Diameter - Microns
VG 7/26/61
359-110

Fig. 5

A # 8

Fig. 6

vg 7/26/61



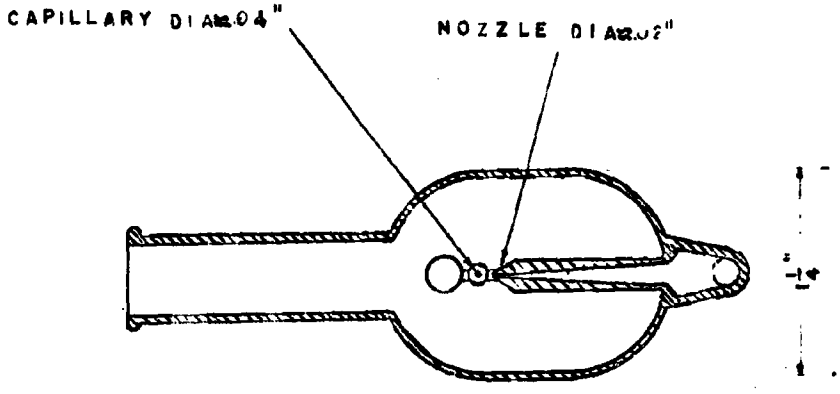


FIG 7 VAPONEFRIN H.E CURRY ALL GLASS NEBULIZER

Fig. 7

K&E 10 X 10 TO THE CM. 359-14
KEUFFEL & ESSER CO. U.S.A.

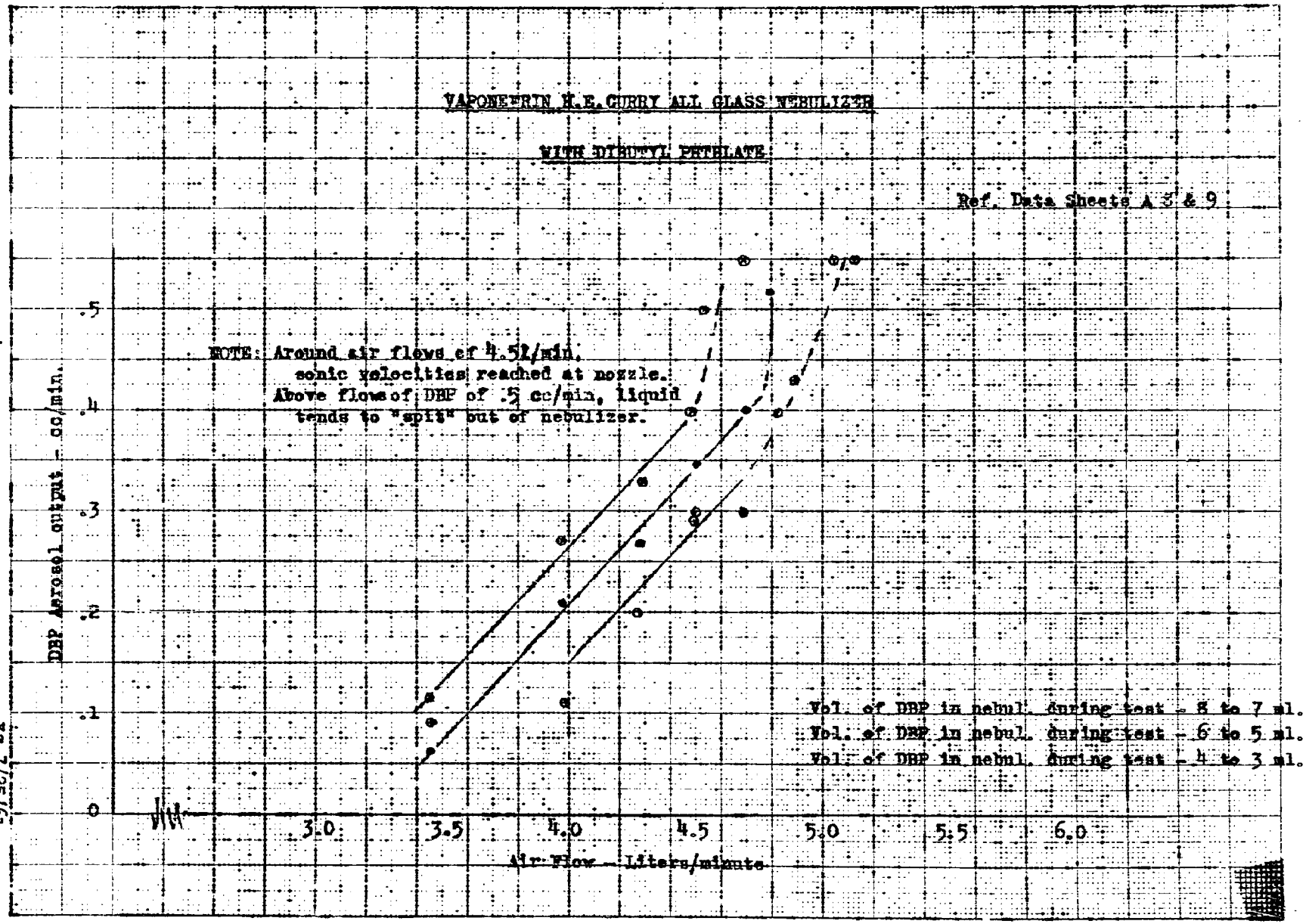


Fig. 8

vs 7/25/61

A # 5

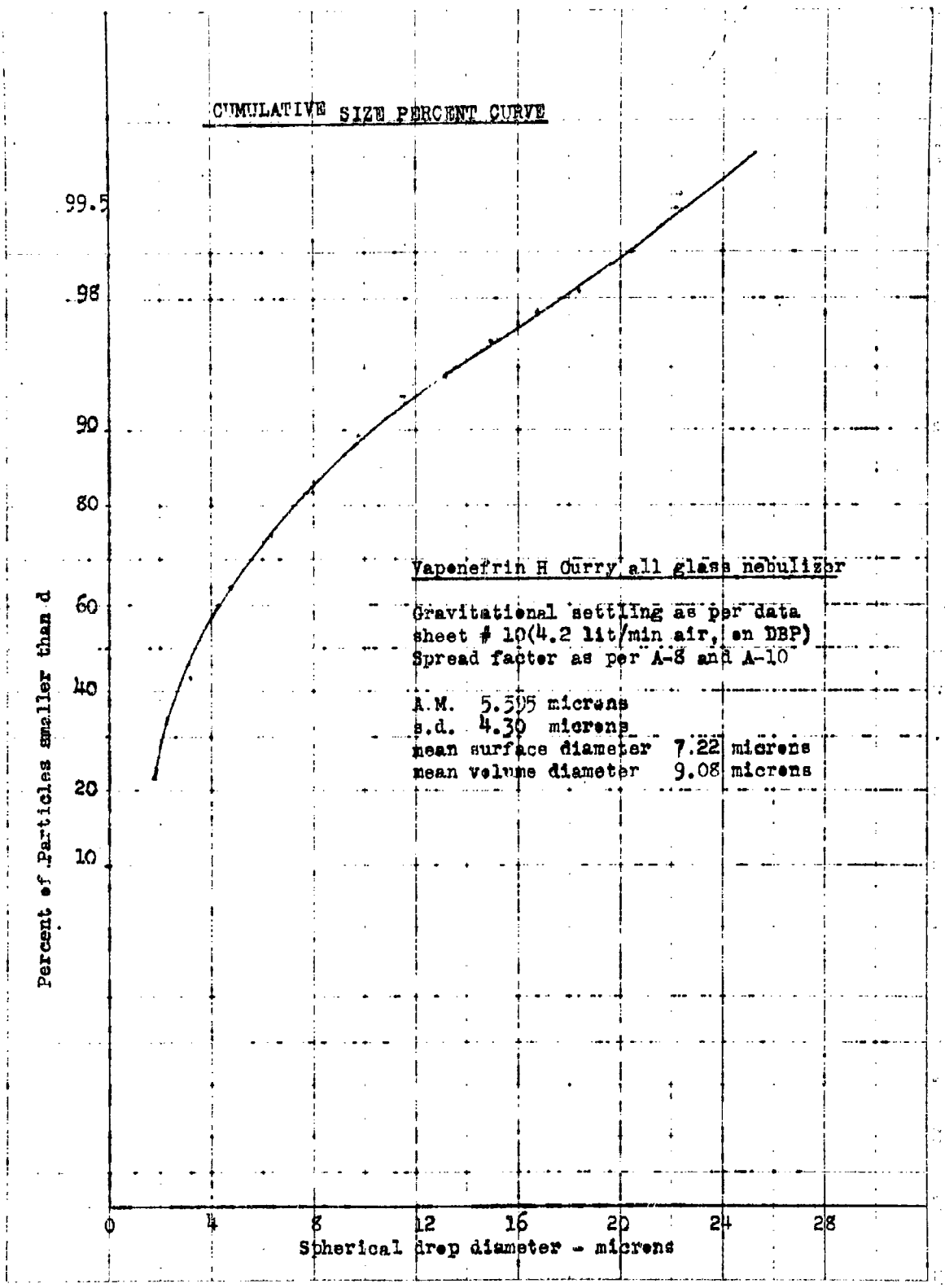


Fig. 9

P.S.# 9
m.c. 8/1/61

AD_____Assesion No._____
Syracuse University, Syracuse, N. Y.
THE SAMPLING OF AEROSOLS IN A TURBULENT AIR FLOW
Progress Report 5, submitted by V. Goldschmidt
The work completed during the fifth period is reported. The La Mer generator has been found inadequate for our test requirements. The output of a nebulizer is evaluated and used for preliminary tests on the use of a hot wire anemometer as a counting device. The relationship of spread factor to drop size for droplets settling with gravitational velocities on clean glass slides is also shown.

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